APS Strategic Planning Meeting

Time-Domain Workshop Summary and Recommendations

Linda Young (ANL) Atomic & Molecular Physics

Lin Chen (ANL) Chemistry & Biology

David Reis (UMich) Condensed Matter Physics

Stephen Milton (ANL) Accelerator Physics

Workshop Chairs

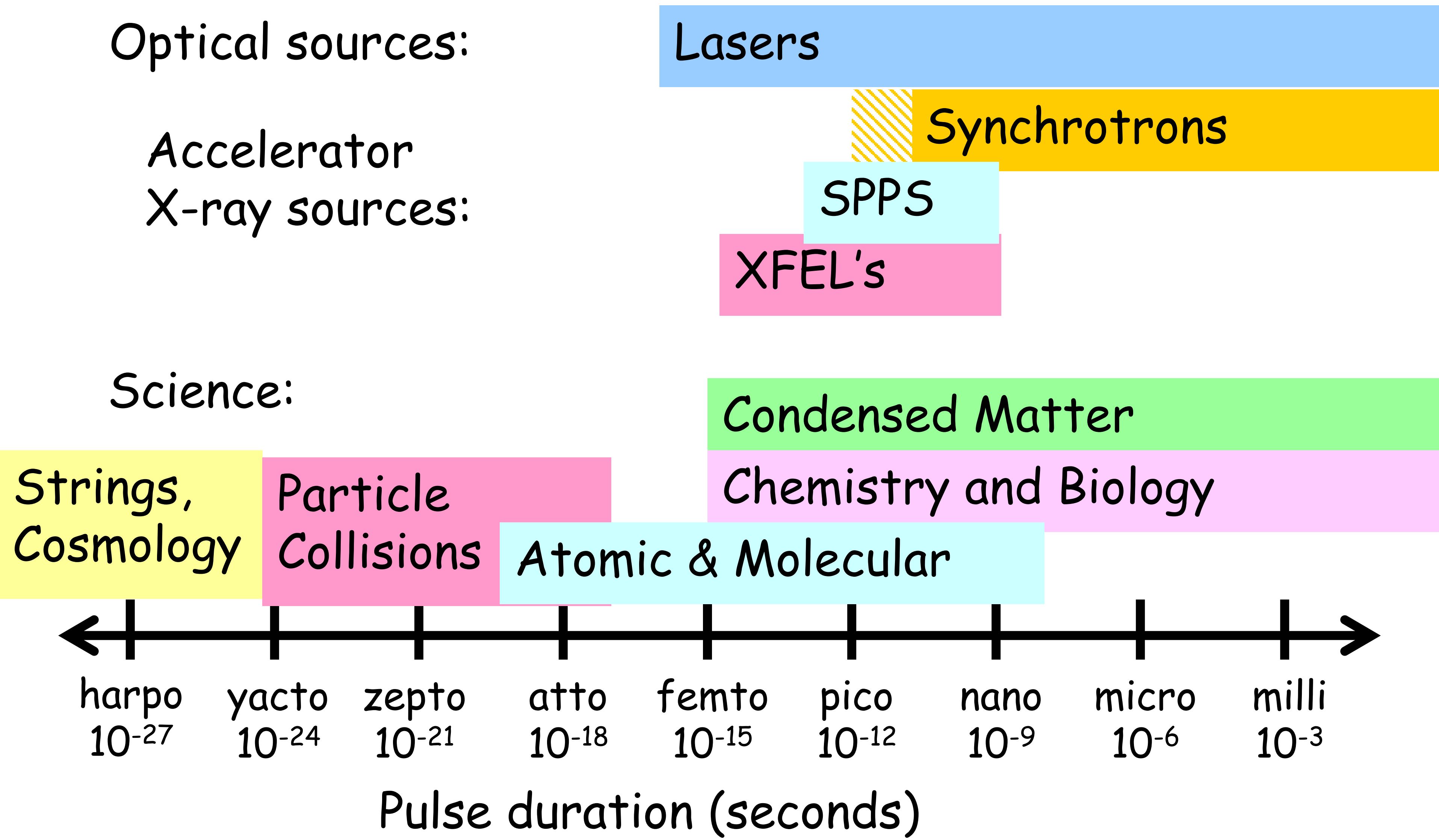
September 2-3, 2004, The Abbey, Fontana, Lake Geneva Area, WI

Workshop Summary and Recommendations

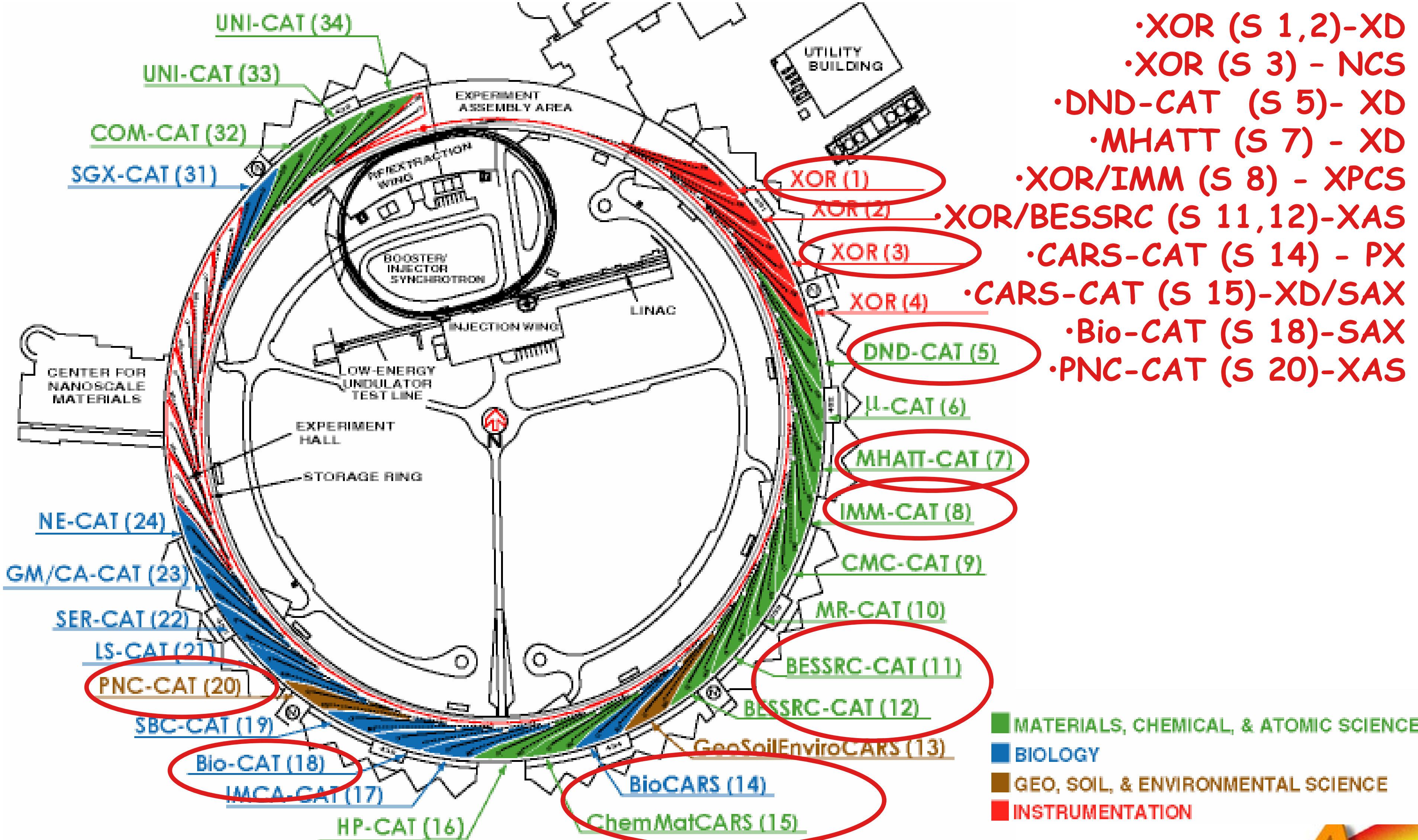
1. ***Facilitate and enhance experiments on timescales ≥ 100 ps***

2. ***Develop short time capabilities of high-flux ≈1 ps pulse at APS***

Ultrafast Sources and Science:



APS Time Domain Science



- XOR (S 1,2)-XD
- XOR (S 3) – NCS
- DND-CAT (S 5)- XD
- MHATT (S 7) - XD
- XOR/IMM (S 8) - XPCS
- XOR/BESSRC (S 11,12)-XAS
- CARS-CAT (S 14) - PX
- CARS-CAT (S 15)-XD/SAX
- Bio-CAT (S 18)-SAX
- PNC-CAT (S 20)-XAS

Scientific Challenges & Opportunities

Atomic and Molecular Physics:
- Understand strong-field effects on inner-shell processes
- Coherent control of molecular processes
- Structural dynamics & phase transitions in isolated targets

Chemical and Biological Dynamics:
- Resolve the fastest time-scale motions of atoms and molecules in order to monitor biological and chemical reactions in real time
- Follow structural evolution correlated to fundamental processes of life and chemistry across multiple timescales
- Explore broad range of molecular dynamics and structural transitions and molecular signaling and energy transduction.

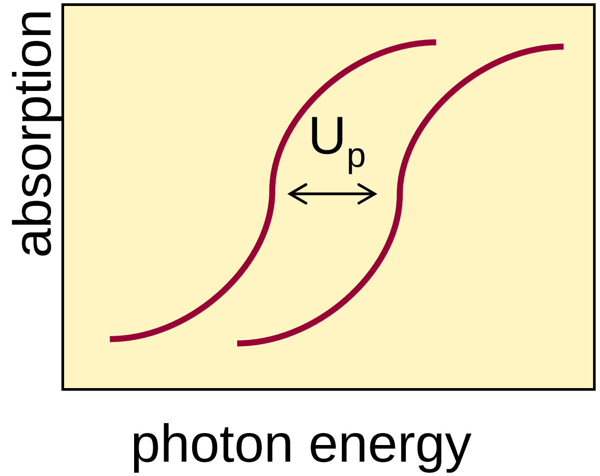
Dynamics in Condensed Matter:
- Nucleation, growth and phase separation
- Nonequilibrium electron and phonon dynamics
- Phase transitions and domain reversals

Ultrafast laser/x-ray interactions: isolated atoms

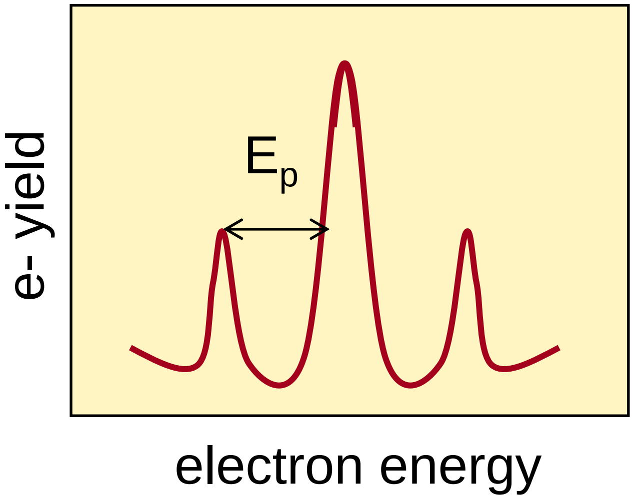
- ***X-ray photoionization is fairly well understood in the weak-field limit***
- ***Understand changes to x-ray processes in presence of strong laser fields***
- ***Theoretical predictions***

  ponderomotive shift in threshold -> absorption spectrum
  free-free transitions in continuum -> electron spectra

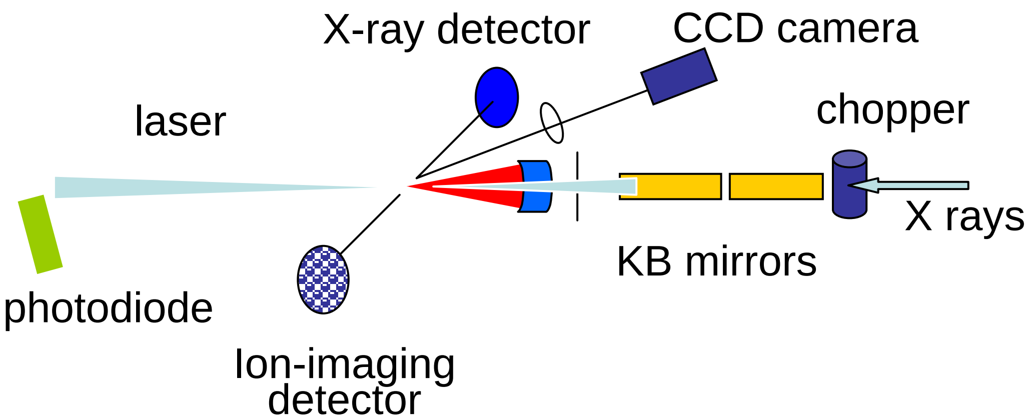
Ponderomotive shift

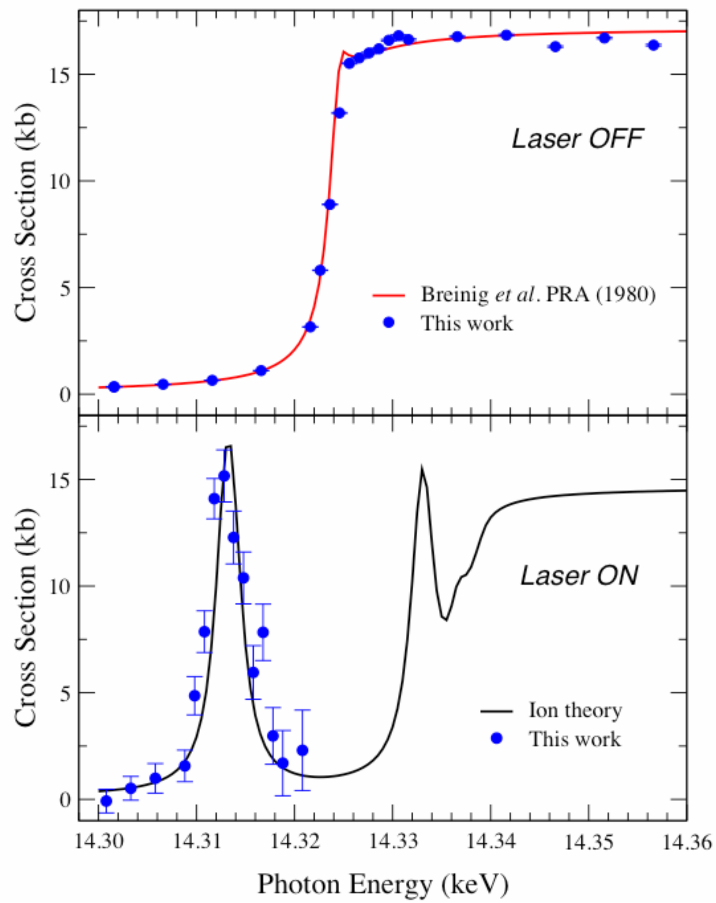


Electron satellites



Laser-modified near-edge spectrum

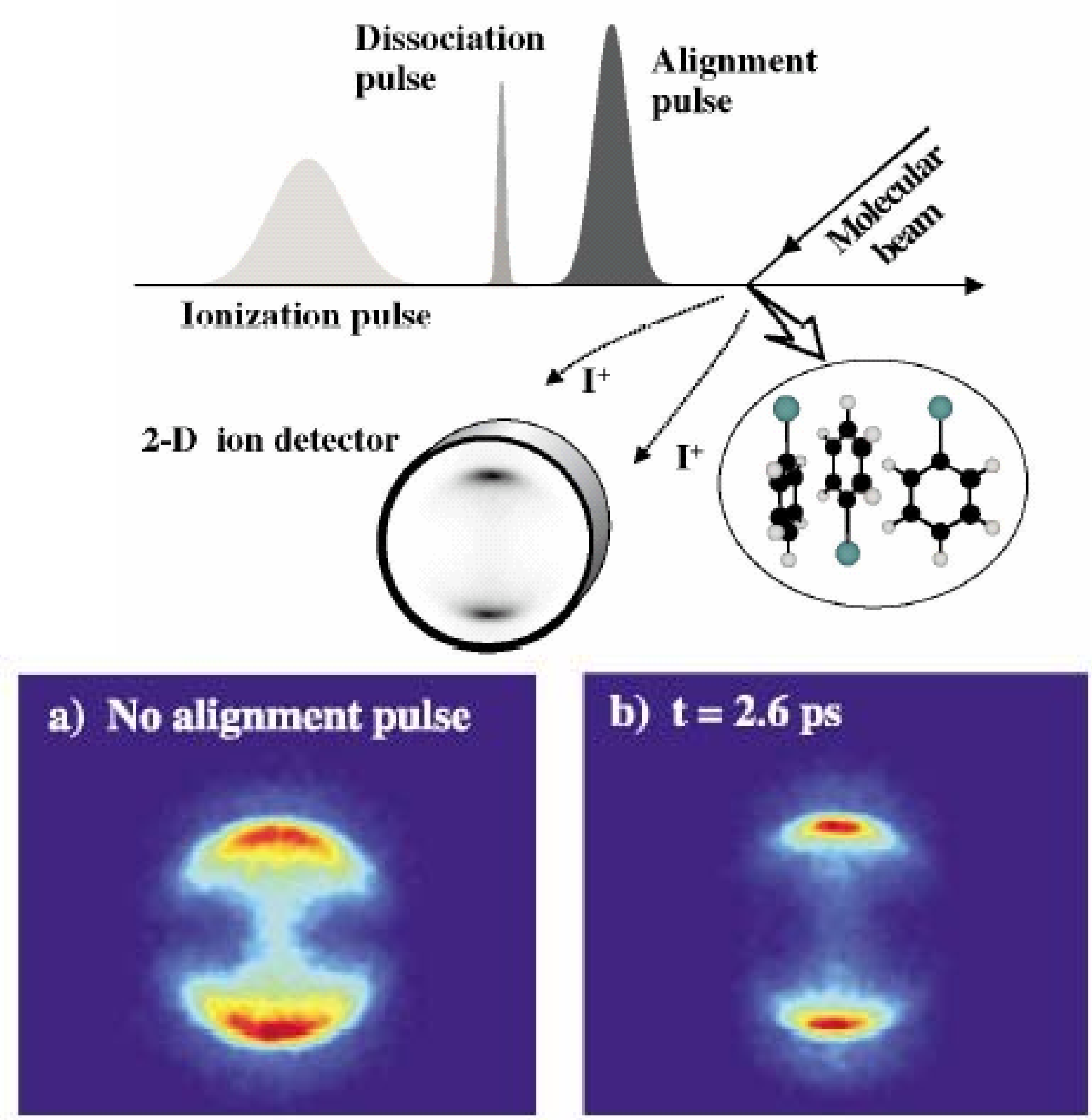




Focusing required for 10^{14} W/cm^2
Laser focused to ≈ 30 µm
X-ray probe central ≈10 µm
Timing and spatial overlap critical

Kr @ ≈ 6 x 10^{14} W/cm^2

Strong-field control of molecular alignment



Transient alignment of molecules

(Here, C_6H_5I viewed by photo-dissociation)

Peronne et al.
Phys. Rev. Lett. **91**, 043003 (2003)

Scientific Challenges & Opportunities

Atomic and Molecular Physics:

- Understand strong-field effects on inner-shell processes
- Coherent control of molecular processes
- Structural dynamics & phase transitions in isolated targets

Chemical and Biological Dynamics:

- Resolve the fastest time-scale motions of atoms and molecules in order to monitor biological and chemical reactions in real time
- Follow structural evolution correlated to fundamental processes of life and chemistry across multiple timescales
- Explore broad range of molecular dynamics and structural transitions and molecular signaling and energy transduction.

Dynamics in Condensed Matter:

- Nonequilibrium electron and phonon dynamics
- Phase transitions and domain reversals
- Nucleation, growth and phase separation

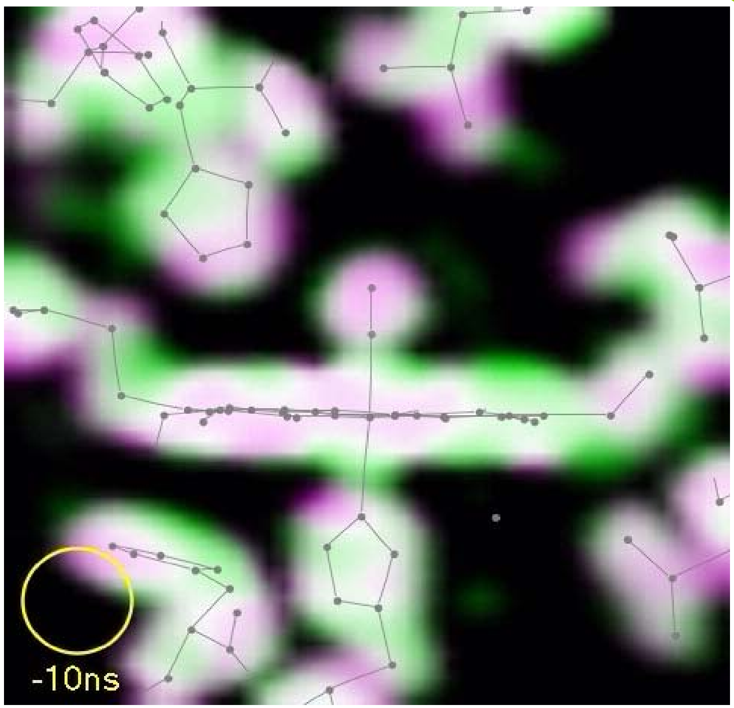
Significance:

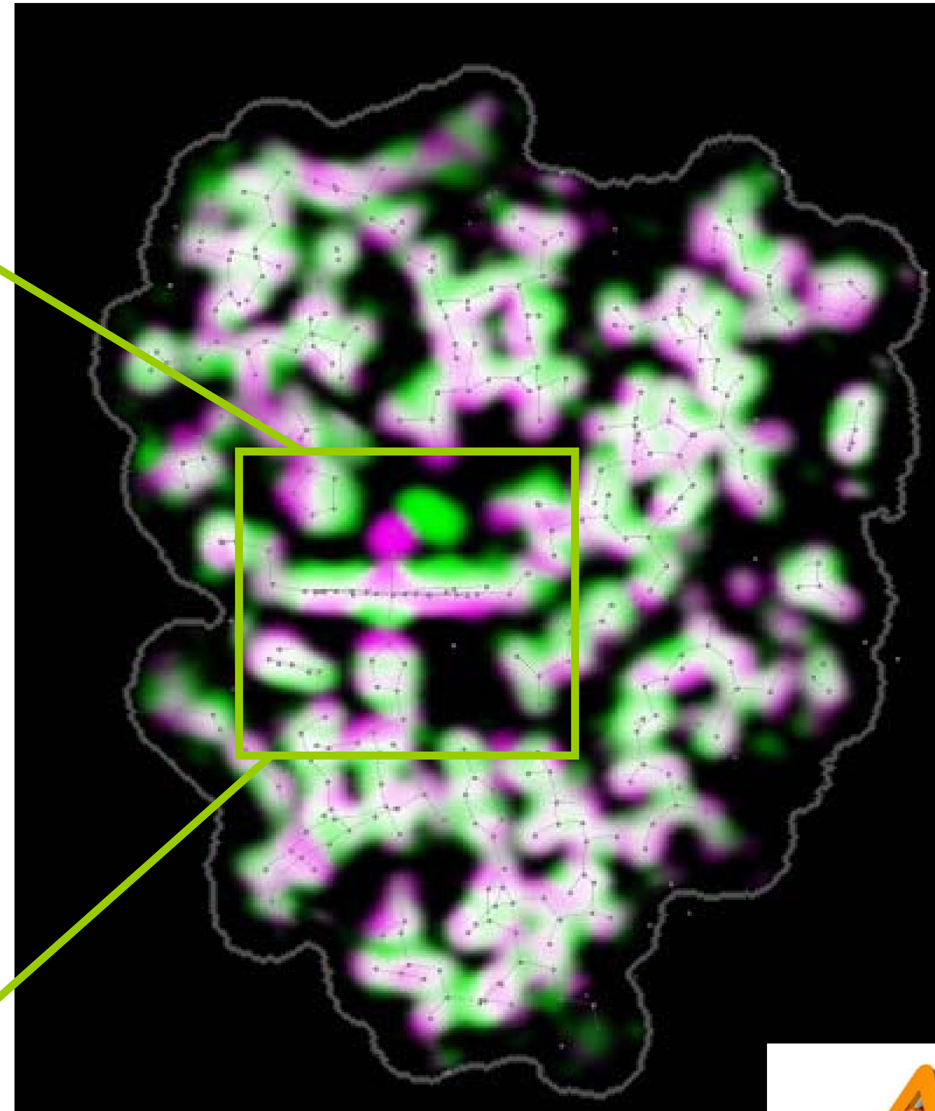
Advances in chemical and biological sciences depend upon the development of correlated time-resolved structural and functional analyses. X-ray techniques provide the most powerful means to resolve molecular structures at the atomic level.

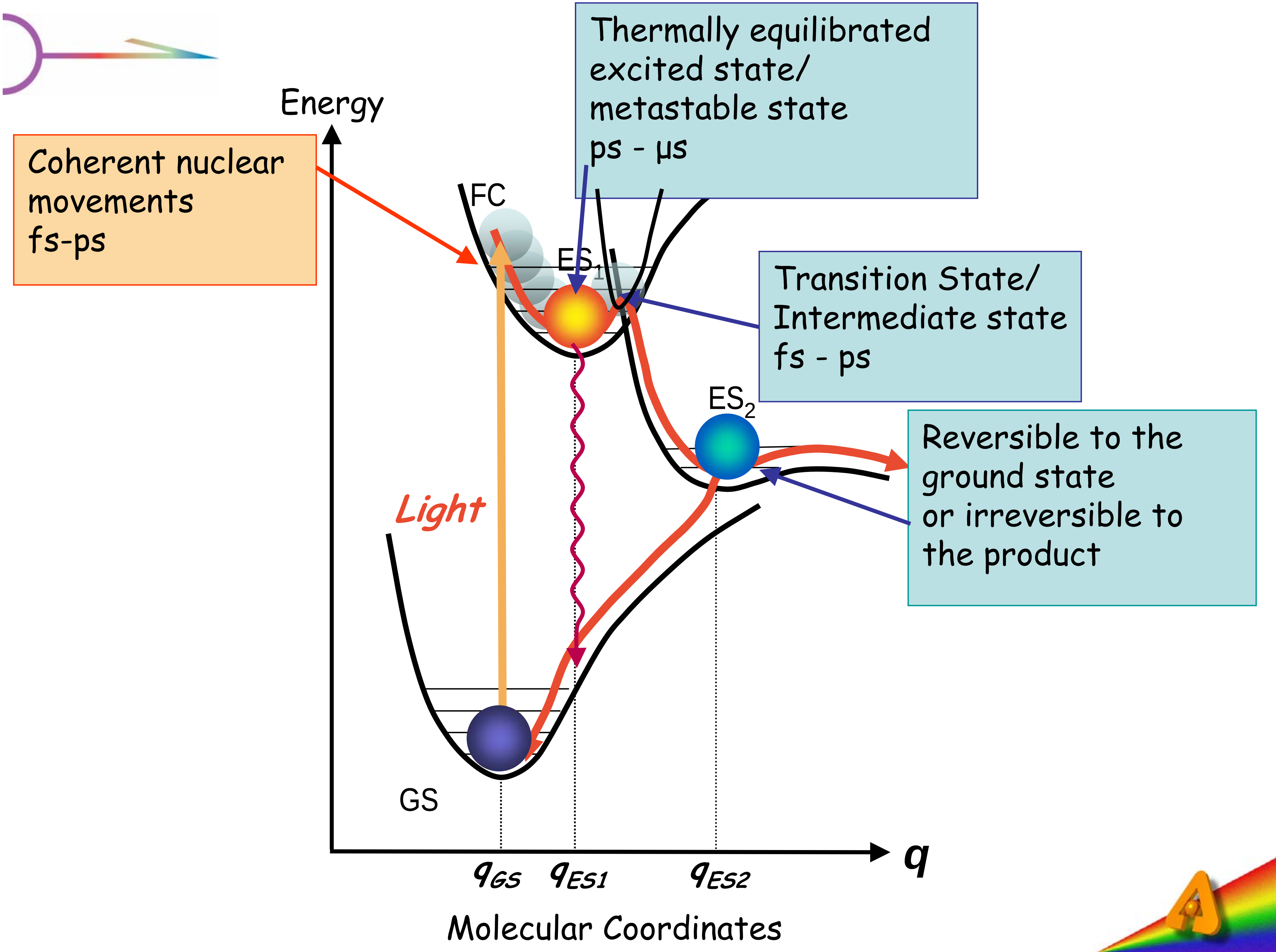
The community of chemists and biologists focused on time-resolved structural studies in chemical and biological sciences is growing fast. Their structural studies are driven by the demands of understanding fundamental chemical and biological processes during the course of reactions. Direct time-resolved structural studies are in the current scientific forefront, and will have a significant impact in a broad range of scientific fields.

Molecular motions in chemical and biological processes take place on multiple time and length scales

Philip Anfinrud et al.
Photodissociation of CO from Myoglobin (ESRF)






Energy
Coherent nuclear movements fs-ps
FC
ES1
Thermally equilibrated excited state/ metastable state ps - µs
Transition State/ Intermediate state fs - ps
ES2
Reversible to the ground state or irreversible to the product
Light
GS
q
qGS
qES1
qES2
Molecular Coordinates

Grand Challenges

Scientific Challenges

- Resolve the fastest time-scale motions of atoms and molecules in order to monitor biological and chemical reactions in real time;
- Follow structural evolution correlated to fundamental processes of life and chemistry across multiple time scales;
- Explore a broad range of molecular dynamics and structural transitions, and mechanisms of molecular signaling and energy transduction.

Operational Challenges

- Effectively utilize unique intrinsic capabilities of APS for time-domain studies to confront the above mentioned scientific challenges;
- Develop infrastructure/technology to make the tools for time-resolved science more broadly available.
- Expand the capacity of time-resolved experimentation at APS

Specific challenges

<u>On the time scale ≥100 ps currently provided by synchrotron x-ray pulses</u>

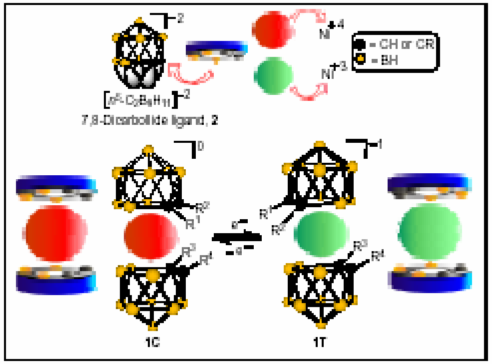
- Extension of time-resolved macromolecular crystallography to other biological systems (light-sensitive or artificially engineered light sensitive, temperature/pressure triggered, fast chemical mixing);

- Time-resolved structural studies need to be extended to
  - Various triggering impulses, including pressure, temperature, electric and magnetic fields for
  - Molecular electronic excited state and reaction intermediate structures;

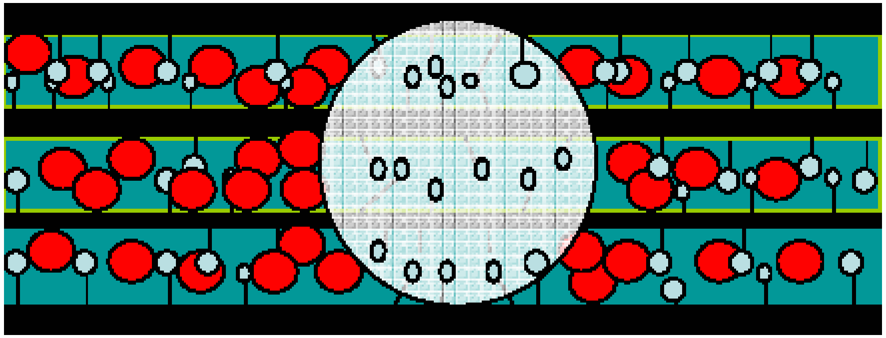
- New direction in time-resolved structural studies includes
  - Structural intermediate in biological enzymatic/protein processes
  - Chemical nanomolecular machines and supermolecules;

MOLECULAR MACHINES

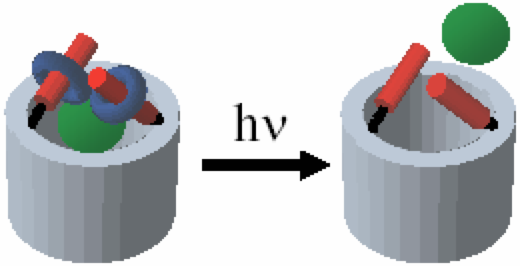
Rotational motion



Moving molecules through pores



Nano valve



M. F. Hawthorne, J. I. Zink, J. M. Skelton, M. J. Bayer, C. Liu, E. Livshits, R. Baer, D. Neuhauser, *Science* 2004, 303, 1849.

Examples

•Structural dynamics in photosynthesis, water-splitting, hydrogen production, solar-energy conversion;

•Atomic level chemical dynamics in condensed phase (solution);

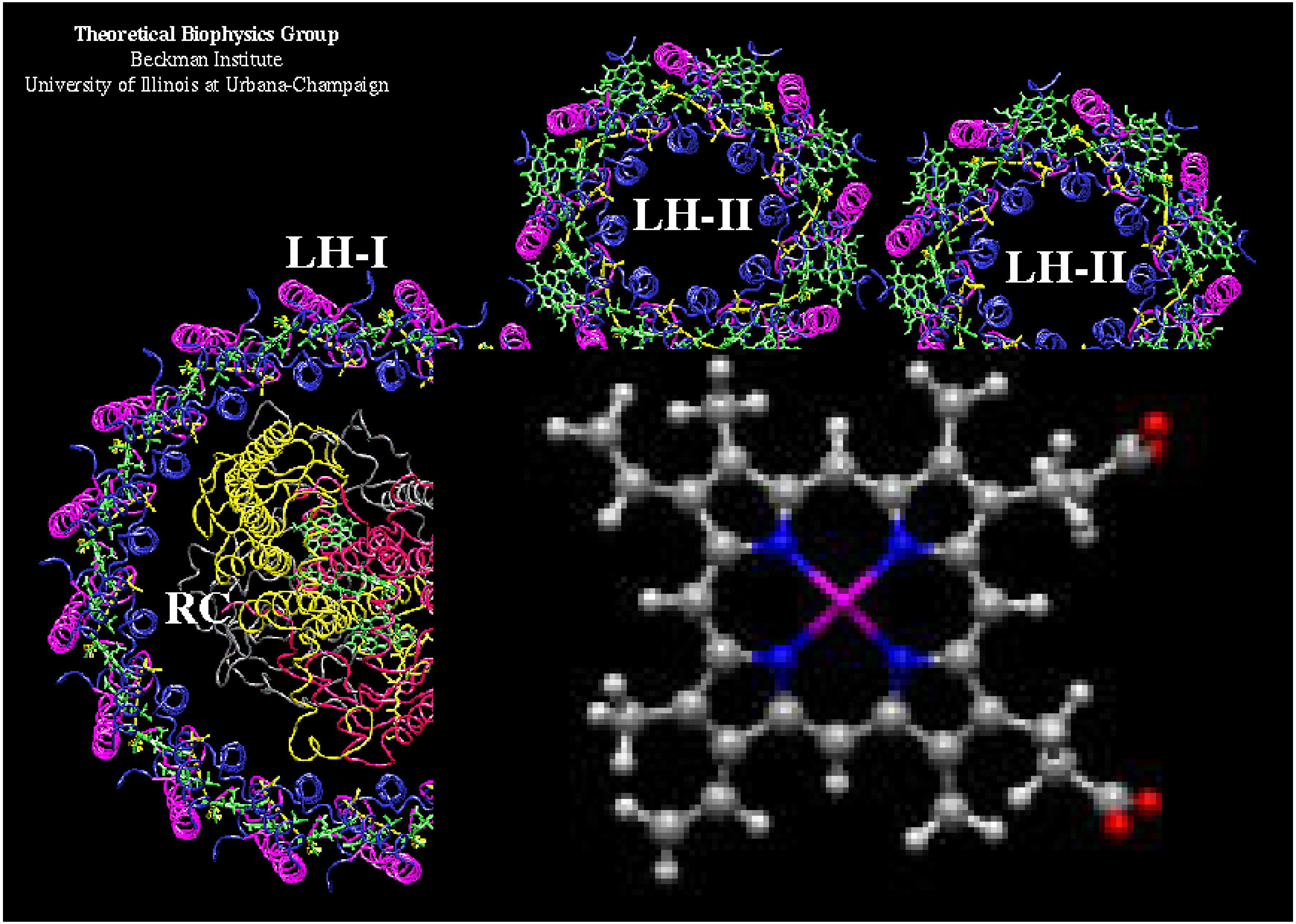
•Solvation processes in chemical reactions;

•Light-driven chemical/biological signal transduction;

•Laser pulse initiated biological and chemical reactions (redox, signaling, molecular switch materials, photocatalysis)

•Dynamics in photochemical reactions of molecules, self-assembled supermolecules, and nanoparticles;

•More detailed considerations of what factors limit experimental accuracy, or the range of samples that can be explored, experiment specific;


Theoretical Biophysics Group
Beckman Institute
University of Illinois at Urbana-Champaign
LH-I
LH-II
LH-II
RC

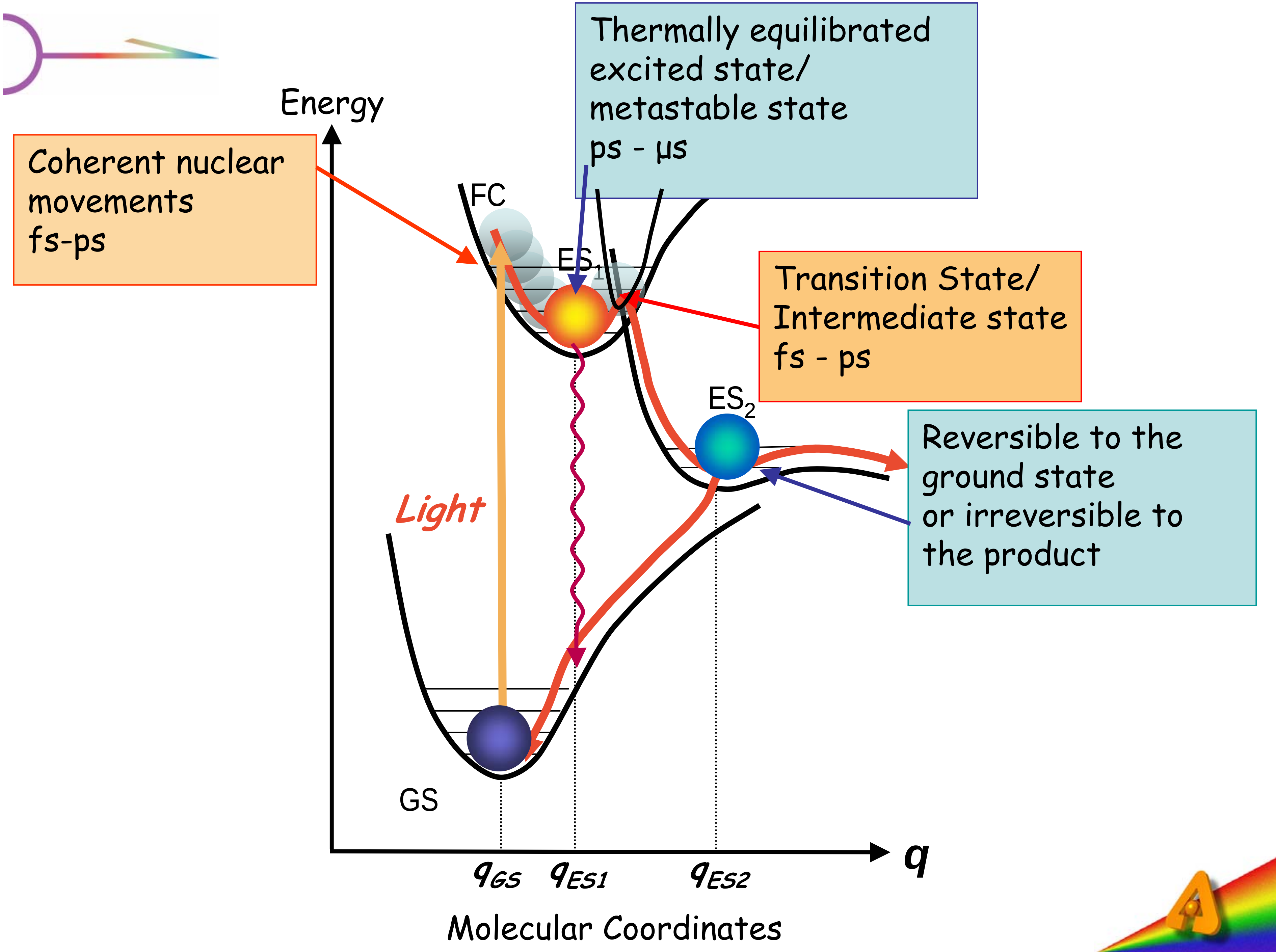
On the time scales much shorter than 100 ps

•Following coherent structural movements on time scale of ~ 1ps from Franck-Condon state through vibrational relaxation to thermally equilibrated excited state;

•Following atomic rearrangements in isomerization, bond breaking/making, electron transfer processes;

•Capturing transition state structures and very short-lived intermediates in catalysis and enzymatic reactions;

•Control of chemical dynamics by diverse character of the “bath” (i.e. nature of the solvent and other media)



Energy
Coherent nuclear movements fs-ps
FC
Thermally equilibrated excited state/ metastable state ps - µs
ES1
Transition State/ Intermediate state fs - ps
ES2
Reversible to the ground state or irreversible to the product
Light
GS
q
qGS
qES1
qES2
Molecular Coordinates

Scientific Challenges & Opportunities

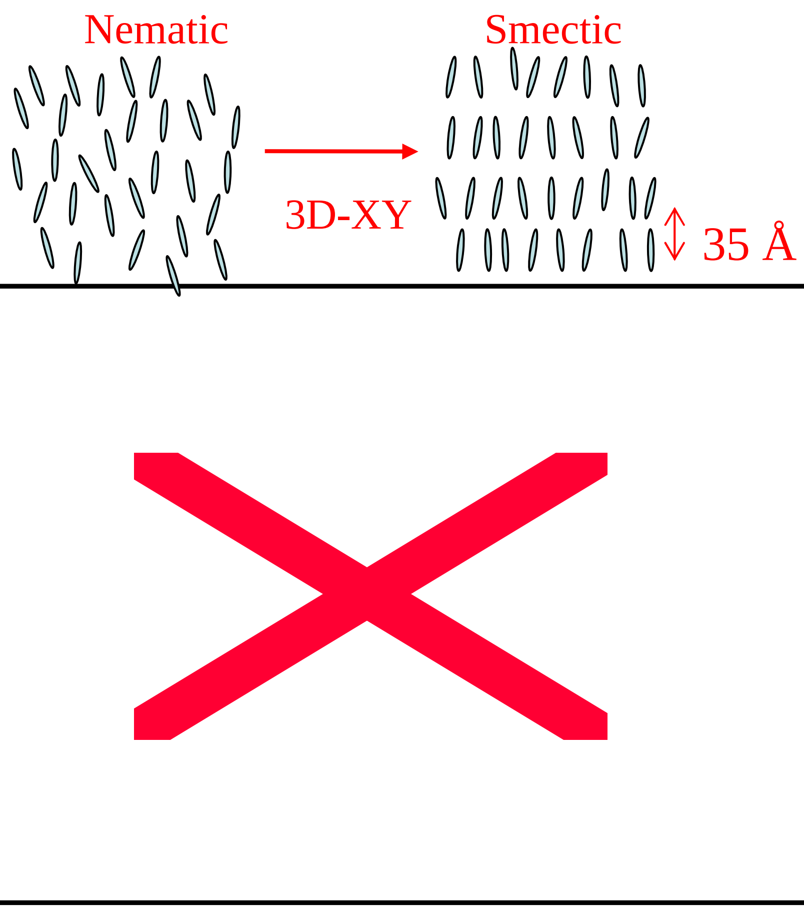
Atomic and Molecular Physics:
- Understand strong-field effects on inner-shell processes
- Coherent control of molecular processes
- Structural dynamics & phase transitions in isolated targets

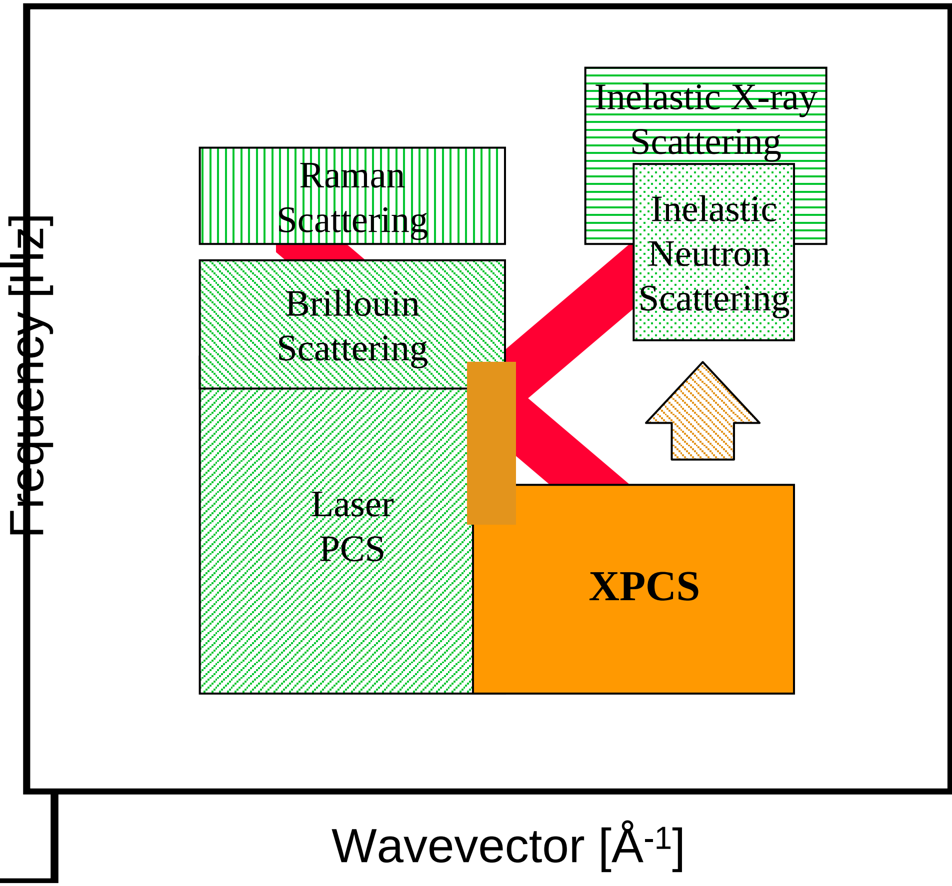
Chemical and Biological Dynamics:
- Resolve the fastest time-scale motions of atoms and molecules in order to monitor biological and chemical reactions in real time
- Follow structural evolution correlated to fundamental processes of life and chemistry across multiple timescales
- Explore broad range of molecular dynamics and structural transitions and molecular signaling and energy transduction.

Dynamics in Condensed Matter:
- Nonequilibrium electron and phonon dynamics
- Phase transitions and domain reversals
- Nucleation, growth and phase separation

Condensed Matter: Coherence Studies

B. Leheny

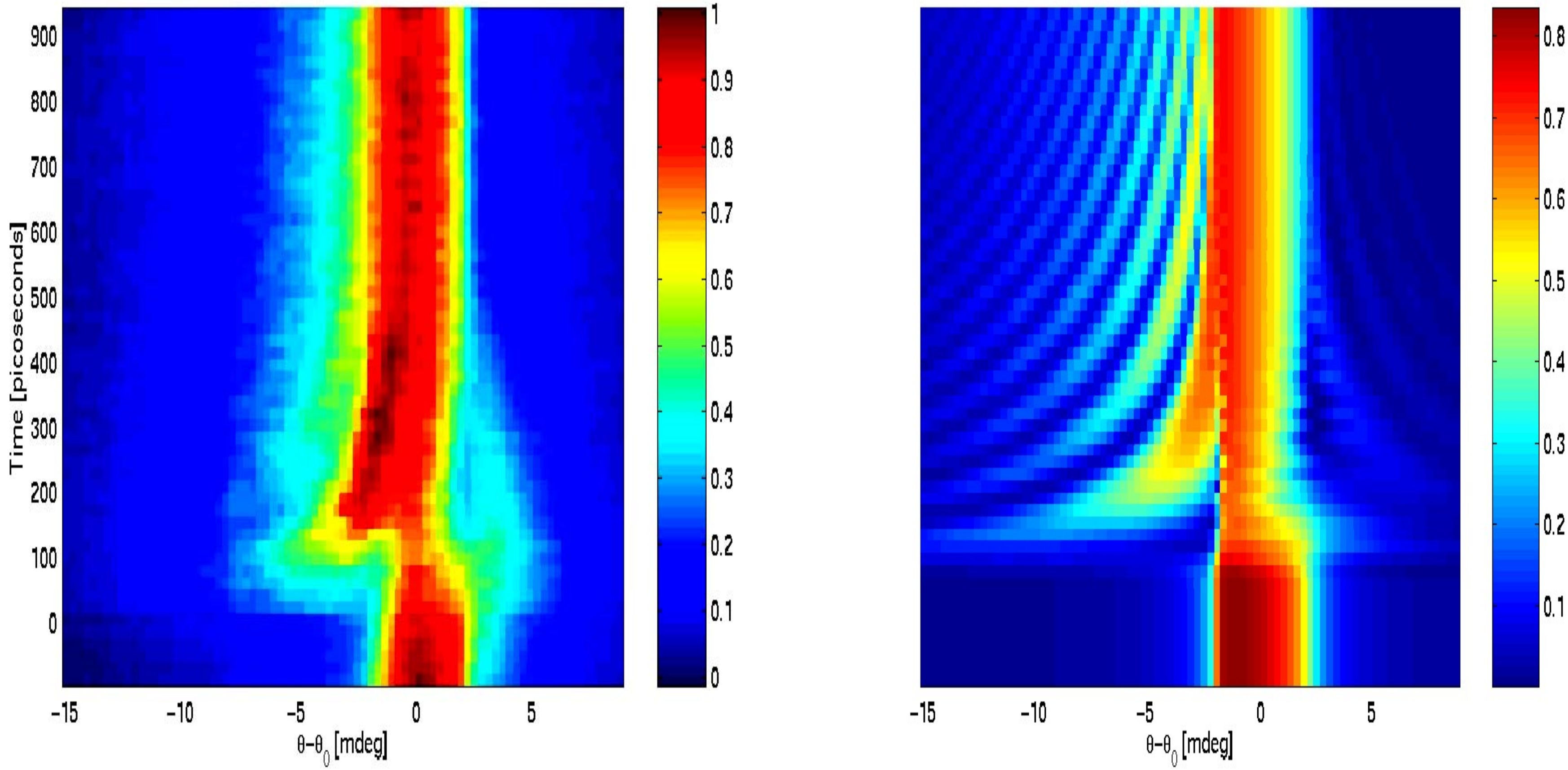




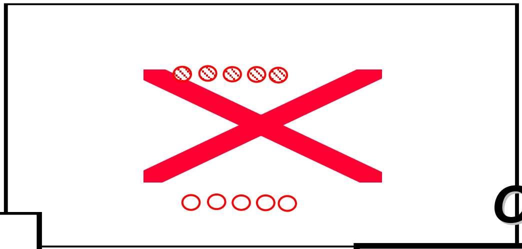
Condensed Matter: Energy relaxation

Time-resolved Bragg Diffraction: Coherent Acoustic Phonons

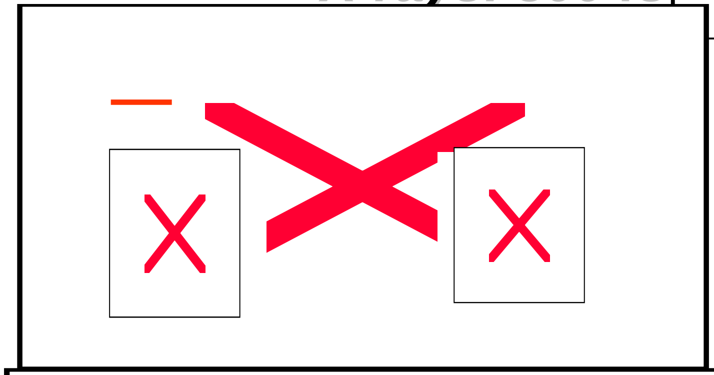
Reis *et al.* Phys Rev. Lett.(86) 2001

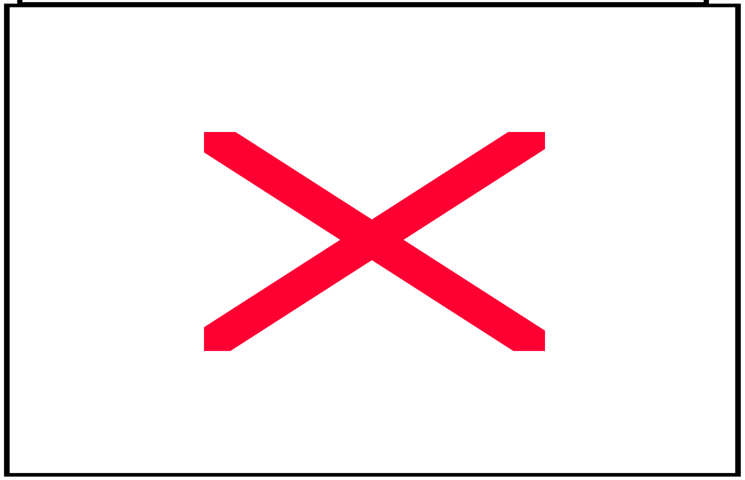


Condensed Matter:
Structural and Electronic Phase Transitions

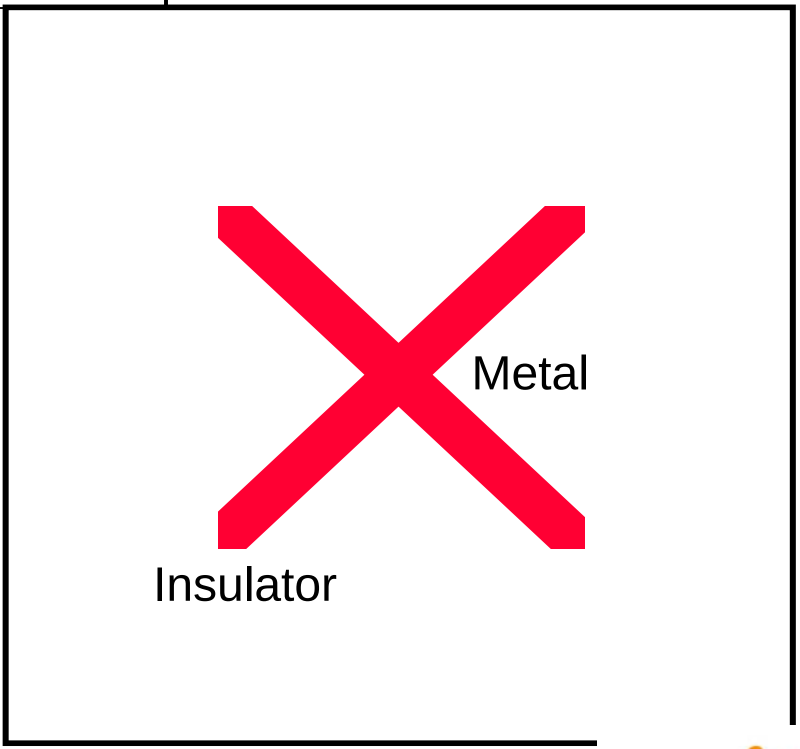


X-rays: 300 fs



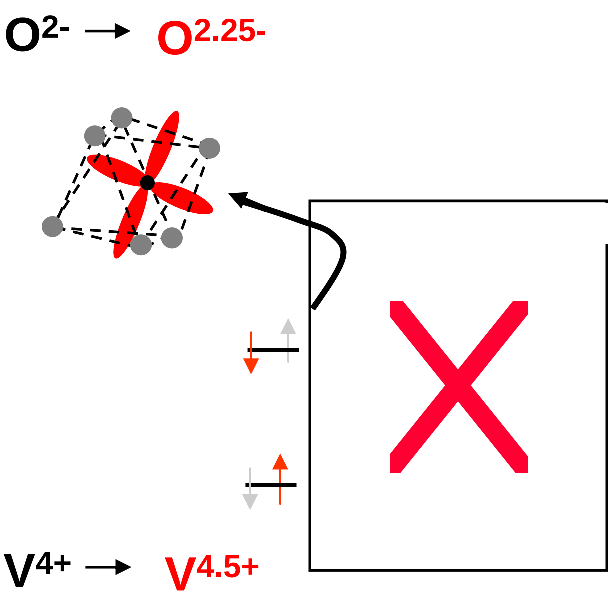


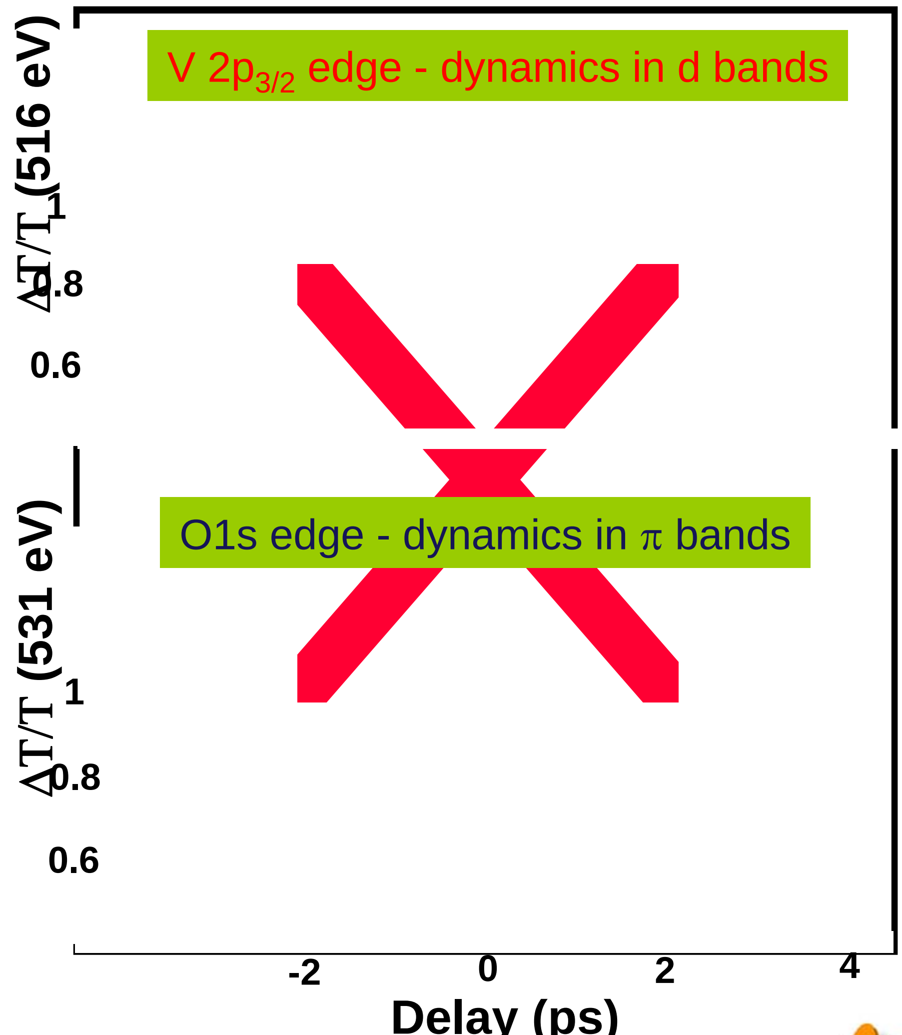
Optical: 100 fs



Cavalleri et al. *Phys. Rev. Lett.* 87, 237401

Shift in Valence?





A. Cavalleri

Workshop Summary and Recommendations

1. Facilitate and enhance experiments on timescales ≥100 ps

2. Develop short time capabilities of APS high-flux ≈1 ps pulse

Achieving Short Pulses in the APS

Possible Methods:

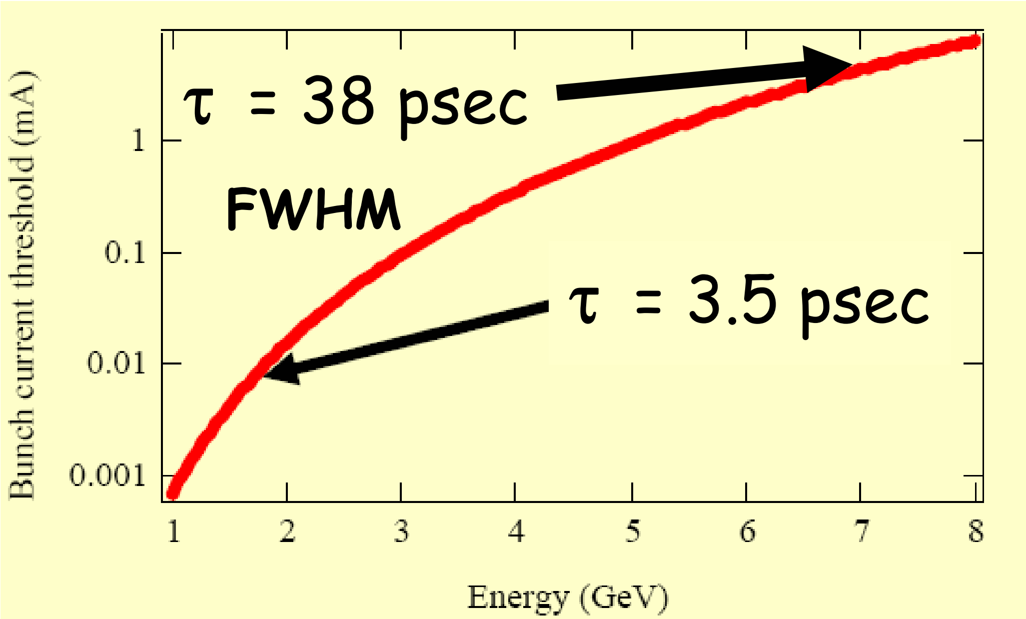
- Equilibrium
  – Limited to Low Charge
- Slicing
  – Limited to low flux
  – Not suitable for the APS
- RF Deflection Method
  – Very Attractive

Bunch Length Limitations

Equilibrium Bunch Length (Zero charge)

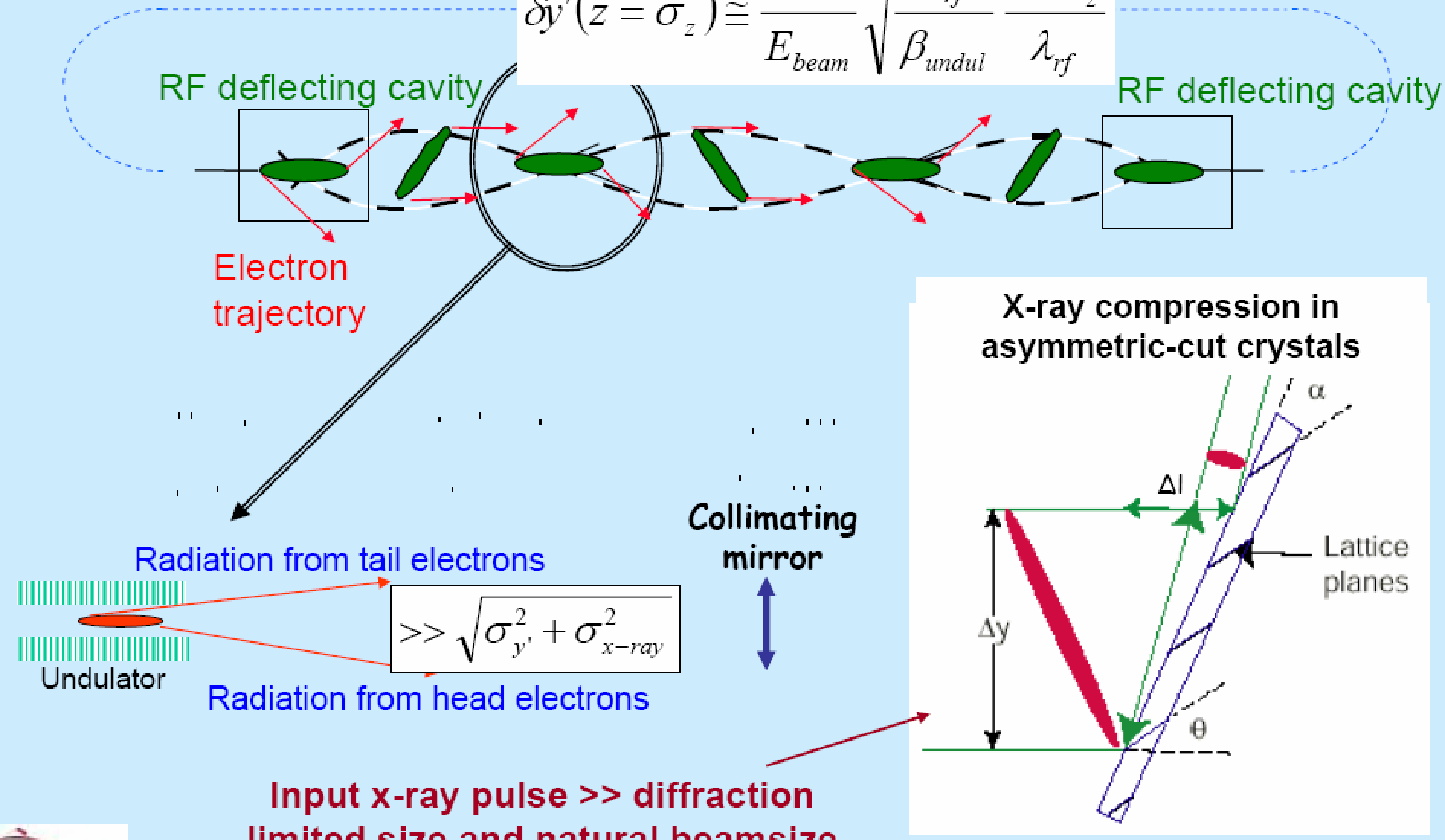
E	7	1.9	3.5	3.5
α	2.3e-4	1.3e-4	1.3e-5	1.3e-4
V_{rf}(MV)	11	22	11	100
σ_τ(psec)	16	1.1	1.25	1.3

Bunch Lengthening Threshold: Best Case



Obtaining short x-ray pulse from a "long" electron bunch

$$\delta y'(z=\sigma_z) \cong \frac{eU}{E_{beam}} \sqrt{\frac{\beta_{rf}}{\beta_{undul}}} \frac{2\pi\sigma_z}{\lambda_{rf}}$$



$$\gg \sqrt{\sigma_{y'}^2 + \sigma_{x-ray}^2}$$

Input x-ray pulse >> diffraction limited size and natural beamsize

Zholents, et al., NIM A 425, pp 385-389 (1999)

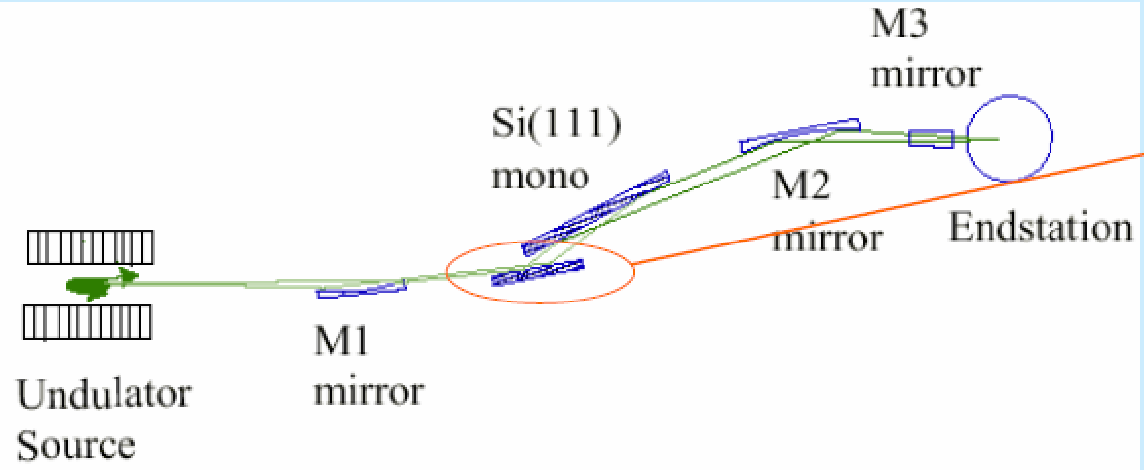
X-ray pulse compression (P. Heimann)

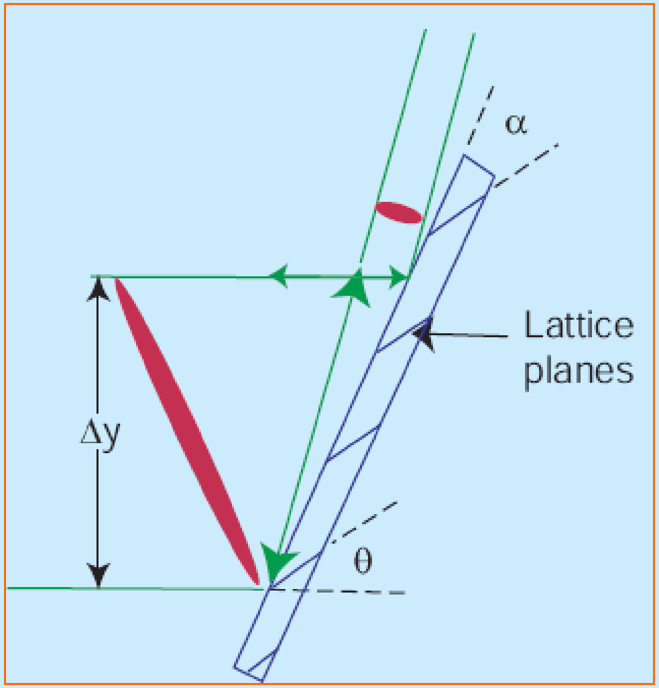
- Optical path length Δl varies linearly with position Δy on crystal

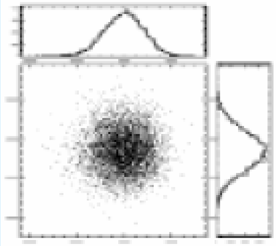
$$\Delta l = 2\ \Delta y \frac{\sin\ \theta\ \ \sin\ \alpha}{\sin\ (\theta + \alpha)}$$

Crystals	λ	Δy	θ	α	Δ*l*
Si(111)	1.5 A	3.8 mm	14.309°	-3.5°	0.6 mm (2 ps)

- We propose to use a pair of asymmetrically cut silicon crystals following collection optics

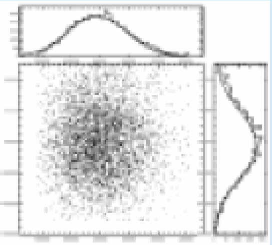






Focus dimensions

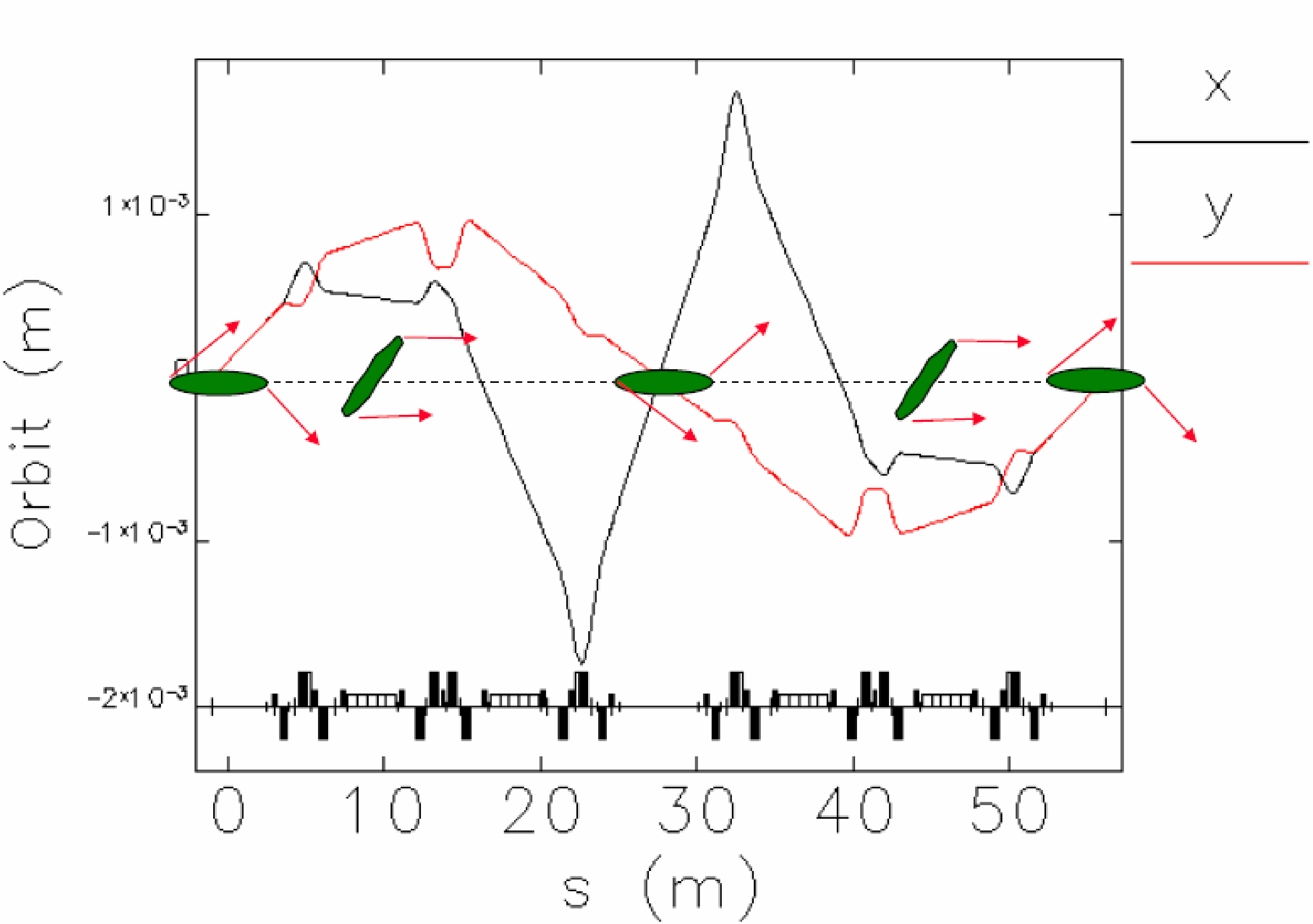
20μm (h) x 12 μm (v)



Focus divergence

1.2 mrad (h) x 500 μrad (v)

Trajectory for an electron with $z=\sigma_z$ and 200 μrad kick



Calculation by Vadim Sajaev, APS Accelerator Physics

Results obtained for undulator beamline:

Beam divergence, $\sigma_{y'}$= 2 μrad

X-ray divergence at 1Å, $\sigma_{r'}$= 3.7 μrad

Total divergence = 4.2 μrad

Total transverse rf voltage = 2 MV

X-ray pulse duration (FWHM) = 2 ps

(compression factor ~ 50)

Results obtained for bend magnet beamline:

Beam size, σ_y= 19 μm

X-ray diffraction size at ω_{cr}=19keV, σ_r= 0.1 μm

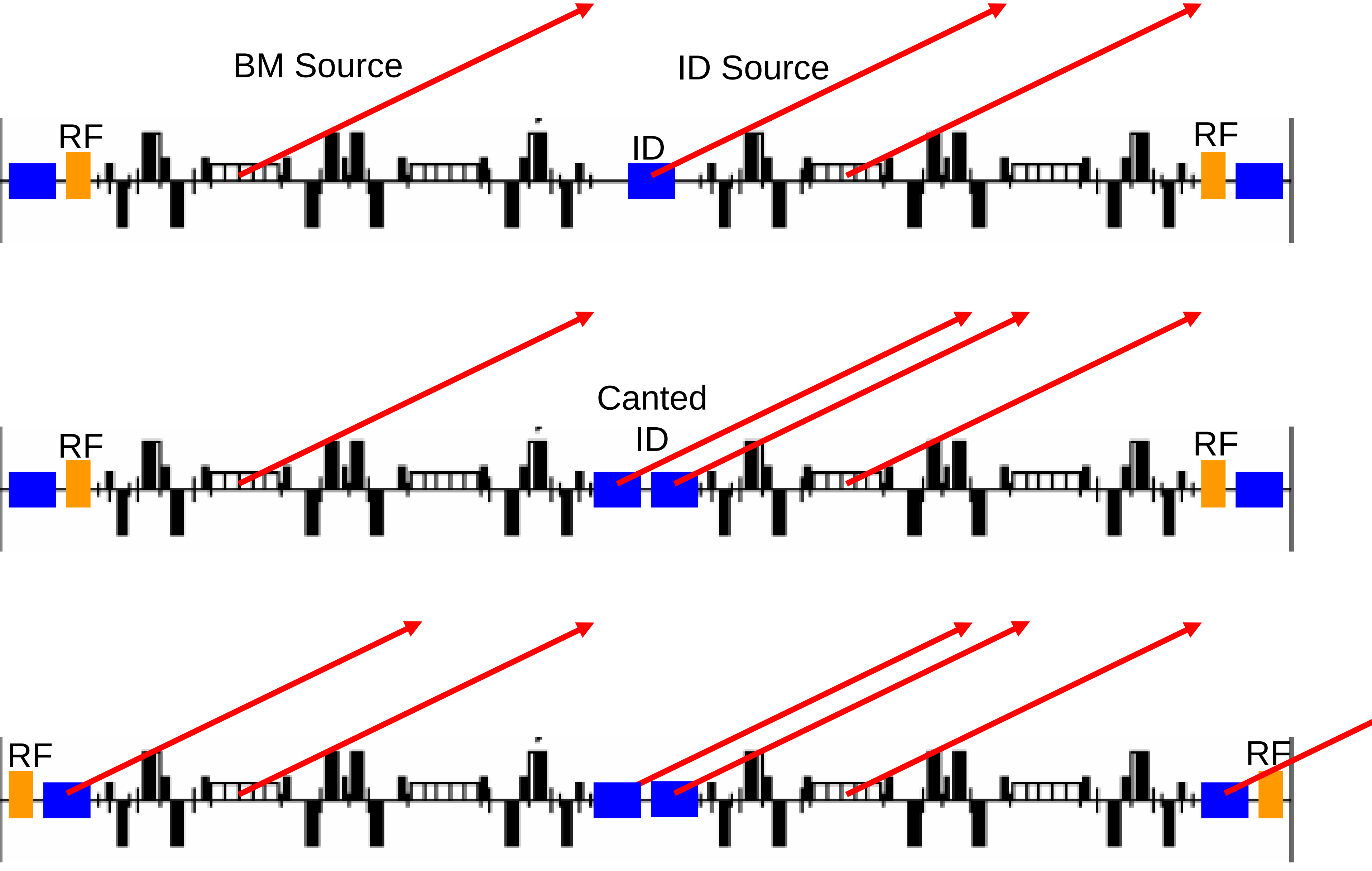
Total size = 19 μm

Total transverse rf voltage = 2 MV

X-ray pulse duration (FWHM) = 1 ps

(compression factor ~ 100)

Various Configurations



Merits of RF Orbit Deflection

- Utilizes x-ray “compression”
  - Avoids charge/bunch dependent issues in the ring
- Full Flux
- Minimal Impact Outside of Region
- Existing (almost) RF Technology
  - Utilized for High Energy Physics Collider “Crab Crossing” Schemes
  - SCRF Deflecting cavity for KEK-B (Japanese B-Factory) exists

Significantly shorter pulses

- Will enable new science
- Will be a unique capability filling “gap” between 100-0.3ps
- Will minimally impact the rest of the ring

Facilitate & Enhance 100 ps capabilities

- ***Ring and beamline operation***
  - Maximize beamtime for time-resolved experiments through suitable choice in standard fill pattern.
  - Optimized insertion devices & optics for timing endstations.
  - Maximize charge in isolated single bunch.
  - Extended x-ray energy range (200-2000 eV).

- ***Technical developments***
  - Advanced chopper → isolate singlet during normal mode
  - Detector development
    - Megapixel direct detection μs readout, unit quantum efficiency
    - Sub-microsecond readout annular detector
    - Sub-picosecond x-ray streak-camera….

Facilitate & Enhance 100 ps capabilities

• ***Management assistance***

Appreciate management support of ultrafast studies through beamline staff hires.

Establish mechanism and funding for joint (APS-PIs) postdoctoral appointees for time-resolved studies.

Time-domain science session at APS user meetings.

Establish time-domain advisory committee for proposal review, bunch pattern operation ...

R & D towards 1 ps capability

- Develop Techniques to Achieve < 100 ps (1 ps goal)
  - –Perform R&D on both accelerator and optics
  - –Seize opportunity to implement on unused sectors of APS